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TEMPERATURE AND MAGNETIC FIELD DEPENDENCE OF CRITICAL CURRENT DENSITY OF YBCO WITH VARYING FLUX PINNING ADDITIONS (POSTPRINT)

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14. ABSTRACT

The critical current density (J_c) of $YBa_2Cu_3O_{7-z}$ films doped with varying flux pinning nanoparticle additions was systematically studied, for applied magnetic fields of H = 0 – 9 T and operation temperatures T = 20 – 77 K. Films were prepared with pulsed laser deposition by $(M/YBCO)_N$ multilayer or $(YBCO)_{1-x}$ M_x single-target methods, for different M phases including Y_2O_3, Y_2BaCuO_5 (Y211) green-phase, and $BaZrO_3$. Very significant differences of J_c (H//c, 20 – 77 K) were measured for optimized M phase additions, that are difficult to model or predict at present. Multilayer films with Y211 and Y_2O_3 nanoparticle additions had the highest J_c (20 – 77 K) for H < 4T and YBCO +BZO-nanorod samples had the strongest J_c (H) for H > 4T and 65 – 77 K, however not for T < 50 K. Seemingly unusual J_c (H,T) properties were measured for $(BZO/YBCO)_N$ multilayer films when compared to YBCO and other doped films; J_c (H) was almost the same as YBCO at 77 K, however at 30 K J_c (H > 2 T) had the strongest properties increasing 70% compared to YBCO +nanoaddition films and increasing 400% compared to YBCO.

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Temperature and Magnetic Field Dependence of Critical Current Density of YBCO With Varying Flux Pinning Additions

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***Abstract*—The critical current density (J_c) of $YBa_2Cu_3O_{7-z}$ films doped with varying flux pinning nanoparticle additions was systematically studied, for applied magnetic fields of $H = 0$–9 T and operation temperatures T = 20–77 K. Films were prepared with pulsed laser deposition by $(M/YBCO)_N$ multilayer or $(YBCO)_{1-x}M_x$ single-target methods, for different M phases including Y_2O_3, Y_2BaCuO_5 (Y211) green-phase, and $BaZrO_3$. Very significant differences of $J_c(H//c, 20-77\ K)$ were measured for optimized M phase additions, that are difficult to model or predict at present. Multilayer films with Y211 and Y_2O_3 nanoparticle additions had the highest $J_c(20-77\ K)$ for $H < 4$ T, and YBCO+BZO-nanorod samples had the strongest $J_c(H)$ for $H > 4$ T and 65–77 K, however not for T < 50 K. Seemingly unusual $J_c(H, T)$ properties were measured for $(BZO/YBCO)_N$ multilayer films when compared to YBCO and other doped films; $J_c(H)$ was almost the same as YBCO at 77 K, however at 30 K $J_c(H > 2\ T)$ had the strongest properties increasing 70% compared to YBCO+nanoaddition films and increasing 400% compared to YBCO.**

***Index Terms*—Critical current density, engineering current density, flux pinning, high temperature superconductor, nanoparticle, $YBa_2Cu_3O_{7-x}$.**

I. Introduction

THE development of high temperature superconductor $YBa_2Cu_3O_{7-\delta}$ (YBCO or 123) thin films on polycrystalline substrates (coated conductors) with a critical current density $(J_c) > 1\ MA/cm^2$ offers great promise for incorporation into power applications such as generators or motors, operating at 40–77 K [1], [2]. YBCO has excellent properties at these temperatures including high engineering critical current density (J_e) in magnetic fields due to strong flux pinning [2], [3]. However, a close examination of the $J_e(H, T)$ properties of commercially available wires [4]–[9], as shown in Fig. 1, indicates the performance is not strong for high-field motor and generator applications requiring $H = 3$ T, which limits the operation temperature to ~40 K. For high power generator and motor applications, the engineering current density is required to reach $J_e > 20\ KA/cm^2$ for 3–5 T fields [9]. The operation temperature is an important factor determining system cost and viability, as the cryogenic efficiency of cryocoolers and vacuum components become steadily worse at reducing temperatures <80 K. For many of these applications, it is preferred to increase the operation temperature >50 K where smaller and more efficient cryocoolers can be utilized. To achieve levels required for motors and generators of $J_e(3\ T) = 20–50\ KA/cm^2$ at temperatures >50 K, Fig. 1 indicates that it is necessary to achieve $J_e(77\ K, self-field) \sim 4 \times 10^4\ A/cm^2$, and possibly even higher to allow a two-times cushion of J_e for flexible operation conditions. This can be achieved by different methods, including increasing I_c in the thin superconducting layers (to >400 A/cm) by increasing the film thickness or using multilayer structures [10], reducing the tape architecture thickness, or increasing $J_{c\ self-field}$. However $J_{c\ self-field}$ of YBCO is almost the maximum that can be achieved by industrial processing methods, and increasing the film thickness or using multilayer structures typically adds additional processing time and cost. One of the most cost-effective methods to increase $J_e(H, T)$ is to strengthen flux pinning, which increases J_c of the film layer substantially in magnetic fields. We will show herein that with addition of optimized nanoaddition pinning centers, it is possibly to consistently increase the operation temperature about 15–20 K or more, for a range of applied field strengths $H > 2$ T.

To increase flux pinning of type-II superconductors such as YBCO, it is effective to introduce a high density of point or correlated defects into the material [2]–[4]. The defects act individually and collectively and to pin the vortex lattice, and different flux pinning models such as weak collective or 3-D strong are used to describe the pinning. The pinning strength of individual defects is a function of the defect size and the interface quality; e.g. $F_p = -dU/dx$, where U is the pinning potential, dU/dx is the derivative as a function of distance, and F_p is the pinning force. Sharp interfaces are desired to maximize F_p. Theoretical studies indicate the defect size should be approximately the coherence length ~2–4 nm at 4.2–77 K to maximize pinning [2], [3].

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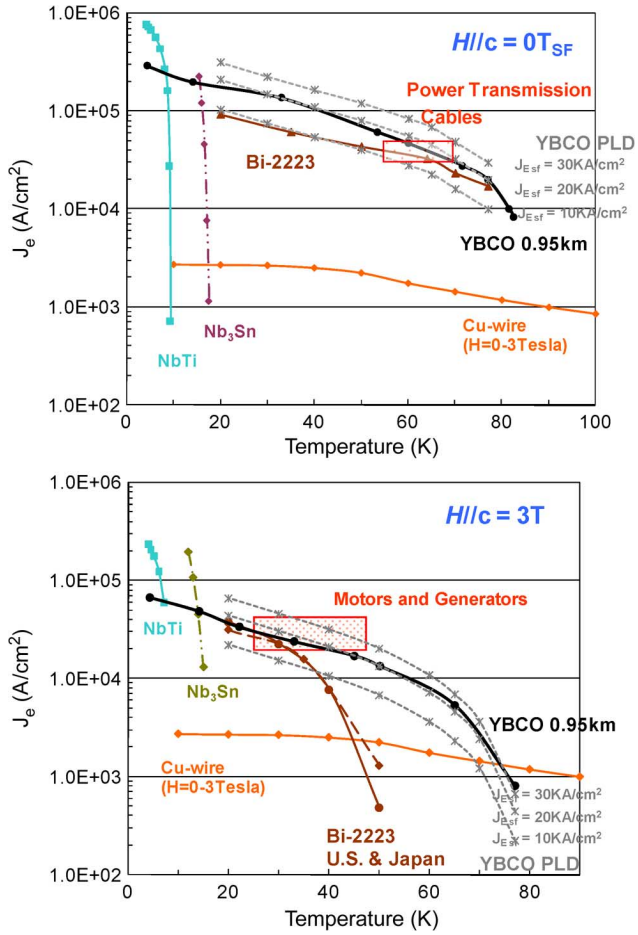

Fig. 1. Engineering critical current density of commercially available ~km length superconducting wires: (top) for $H = 0\ \mathrm{T_{self-field}}$, and (bottom) for $H = 3\ \mathrm{T}$. Dependence of J_e on temperature is from [4]–[9]. Goals for J_e are from [9], and temperature ranges are estimated for operation of BSCCO or YBCO wires. Dashed lines estimate the temperature dependence for different $J_e(77\ \mathrm{K, self-field})$ values of YBCO films deposited by PLD onto buffered-Ni-alloy substrates with 1 micron thickness and 0.04 cm architecture, and conservatively assuming $J_{ct}(77\ \mathrm{K, self-field}) = J_{cm}(77\ \mathrm{K, self-field})$. The values of $J_e(H, \mathrm{T})$ are remarkably close comparing commercial YBCO 0.95 km wires and those estimated for PLD of YBCO.

Many methods of introducing flux pinning defects into YBCO have been considered [9]–[23], including addition of nanoparticles by multilayer [11]–[19] and single-target methods [9], [10], [20]–[23], and other methods [10], [23]. For multilayer $(\mathrm{M_n/YBCO_m})_N$ films, a number of different second-phase additions M have been studied including $\mathrm{M} = \mathrm{green-phase\ Y_2BaCuO_5}$ (Y211), $\mathrm{Y_2O_3}$, $\mathrm{CeO_2}$, $\mathrm{IrZrO_3}$, and other phases with negative effects on T_c and/or J_c(H) including $\mathrm{M} =$ La211, MgO, and Sm123 [11]–[19]. For $(\mathrm{RE123})_{1-x}\mathrm{M_x}$ single-target films, a number of materials have been considered that provided increases of flux pinning including $\mathrm{M} = \mathrm{BaZrO_3}$ and $\mathrm{BaSnO_3}$ and minute dopants such as Tb, Nd, and Pr [9], [10], [20]–[23].

Herein studies the $J_c(H, \mathrm{T})$ properties of selected YBCO+nanoaddition films prepared by both multilayer and single-target pulsed laser deposition. While there are many recent papers published with strong increase of flux pinning, it is difficult to compare results of different publications for some types of nanoadditions because of many experimental variables including film thickness, processing parameters, and varying substrates including single crystal and buffer-coated metallic substrates [9]–[23]. In this paper, to make useful comparisons of different nanoadditions, experimental conditions for pulsed laser deposition were kept almost constant to make consistent comparisons.

II. Experimental

Multilayer $(\mathrm{M/YBCO})_N$ and $(\mathrm{YBCO})_{1-x}(\mathrm{BZO})_x$ films were deposited by pulsed laser deposition (PLD), using parameters and conditions described in detail previously [11], [12], [17], [24]. Herein, for $(\mathrm{YBCO})_{1-x}\mathrm{M_x}$ single-target films x will denote volume %. Deposition parameters were 248 nm laser wavelength, 3 $\mathrm{J/cm^2}$ laser fluence, 25 nm pulse length, 4 Hz laser repetition rate, 5.5 cm target-to-substrate distance, heater block temperature 825°C for $\mathrm{Y_2O_3}$ and Y211 and 800°C for $\mathrm{M} = \mathrm{BZO}$, 83–90% dense targets, 300 mTorr oxygen partial-pressure, and a post-deposition anneal at 500°C and 1 atmosphere of oxygen [12], [24]. Substrates were $\mathrm{LaAlO_3}$ (LAO) and $\mathrm{SrTiO_3}$ (STO) 100 oriented single crystals, with epi-polish.

The $(\mathrm{M/YBCO})_N$ multilayer films were first optimized by varying the M layer thickness [17]. An optimal surface layer coverage corresponding to M phase <1 nm thickness was found to be necessary to increase $J_c(H)$ compared to YBCO. The optimal layer thickness for each M phase was kept constant in this experiment: $\mathrm{Y_2O_3} \sim 0.5$ nm, Y211 ~ 0.8 nm, and $\mathrm{BaZrO_3} \sim 0.5$ nm [17]. Using the optimal M phase thickness, the YBCO layer was also systematically varied for $(\mathrm{M/YBCO})_N$ multilayer films and optimized for maximum $J_c(65\ \mathrm{K}, 3\ \mathrm{T})$. The details of this study will be shown elsewhere, however characterization of the best films from this optimization study are presented here. Optimization of $(\mathrm{YBCO})_{1-x}(\mathrm{BZO})_x$ films was studied on single crystal substrates for the experimental conditions here previously for $\mathrm{BZO} = 0$–2 Vol % [19], however extended to $\mathrm{BZO} = 4$–6 Vol % [25]. Properties of the films are shown in Table I. Film thicknesses were in the range of 0.2 to 0.3 μm. The T_c transition widths were all narrow about 1.5 K for T_c FWHM, as measured by AC susceptibility with $h = 2.2$ Oe sensing field [11], [12].

III. Results and Discussion

Critical current densities of optimized films are shown in Fig. 2, for varying temperature and magnetic field. The $(\mathrm{M/YBCO})_N$ multilayer films doped with $\mathrm{M} = \mathrm{Y_2O_3}$ and Y211 nanoparticles have almost the same $J_c(H, \mathrm{T})$ dependence for all temperatures and fields studied, and have the strongest increase of $J_c(30\text{–}77\ \mathrm{K})$ for $H < 3$–4 T of any nanoadditions compared thus far. At 65–77 K the J_c increase compared to YBCO is up >300% for $H < 3$ T, and $J_c(H)$ is significantly strongly than obtained with YBCO doped with $\mathrm{BZO} = 2$–4 Vol % BZO nanorods. It's somewhat puzzling why the BZO nanorod additions have smaller pinning strength than Y211 or $\mathrm{Y_2O_3}$ nanoparticle additions, since the BZO nanorods have almost nearly perfect size and shape. One might expect the pinning performance to be stronger at all H-fields for BZO nanorod additions, compared to Y211 or $\mathrm{Y_2O_3}$ nanoparticle

TABLE I
PROPERTIES OF YBCO+NANOPARTICLE FILMS

Film	M Phase Layer Thickness (nm)	YBCO Layer Thickness (nm)	Nanoparticle Vol%	T_c(K)
YBCO			0	91.0
$(Y_2O_3/YBCO)_{35}$	0.5	6.5	6.8	89.8
$(Y211/YBCO)_{35}$	0.6	6.7	8.6	89.7
$(BZO/YBCO)_{52}$	0.5	5.4	9.1	88.1
$(YBCO)_{0.98}(BZO)_{0.02}$			2	87.8
$(YBCO)_{0.96}(BZO)_{0.04}$			4	87.0

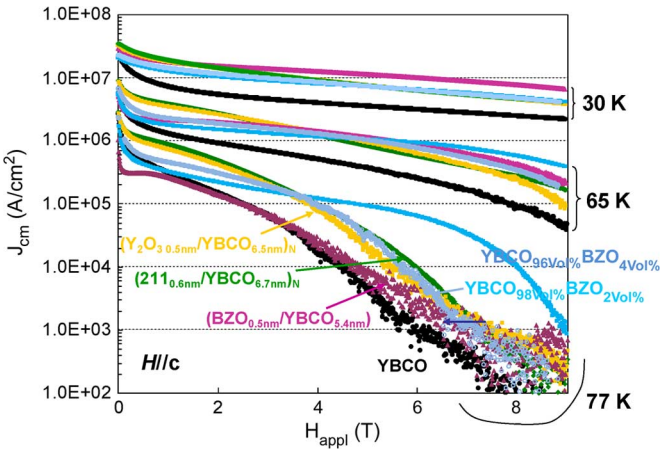

Fig. 2. Magnetic critical current density as a function of applied magnetic field for YBCO+nanoparticle films, measured at 30 K, 65 K and 77 K.

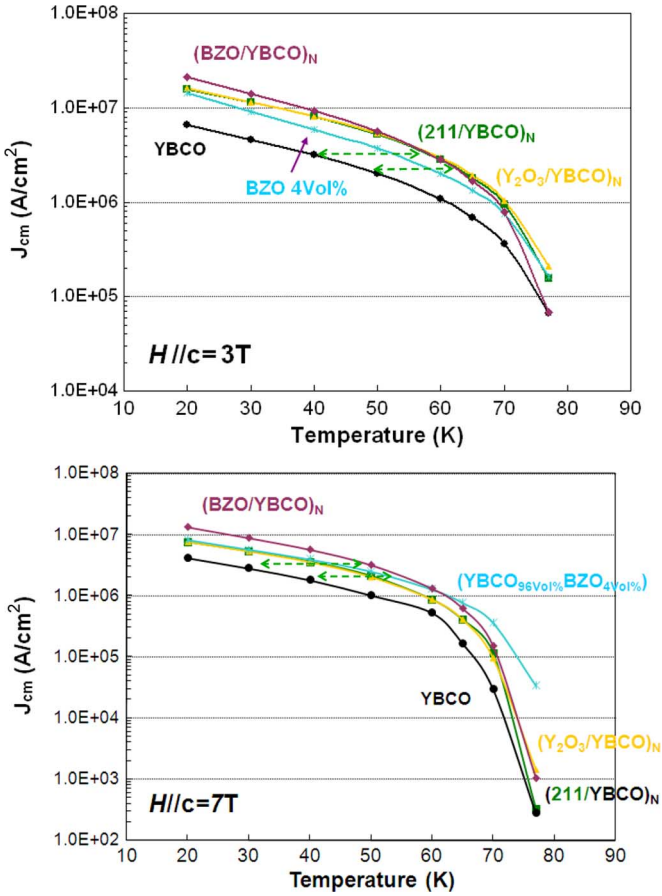

Fig. 3. $J_{\rm cm}$(H = 3 T) plotted as a function of temperature for YBCO+nanoparticle films: (top) $H = 3$ T, (bottom) $H = 7$ T.

additions, however that is not the case. The comparisons are consistent at lower temperatures of 30 K to 65 K, showing Y211 and Y_2O_3 nanoparticle additions have stronger flux pinning than BZO nanorods at $H < 4$ T. However for $H > 4$ T, the $(YBCO)_{1-x}(BZO)_x$ single target samples compared to $(M/YBCO)_N$ multilayer films are clearly superior for 77 K and 65 K, however are the same at 30 K.

The $(BZO/YBCO)_N$ multilayer films demonstrate very interesting and unusual $J_c(H, T)$ properties, especially when compared to YBCO and other YBCO+nanoaddition films. At 77 K, the $J_c(H)$ dependence is almost the same as for YBCO; e.g. the film quality is the same or worse when J_c is considered. However at 65 K the $(BZO/YBCO)_N$ film has comparable $J_c(H)$ properties to the other YBCO+nanoparticle films, and at 30 K it has the highest $J_c(H)$ properties of any of the films. The $(BZO/YBCO)_N$ films have the highest $J_c(30\ K)$ for $H > 3$ T and the increase of J_c is ~70% for $H > 8$ T compared to other YBCO+nanoparticle films, and almost 400% when compared to YBCO. A similar unusual temperature dependence comparing 77 K and 65 K was also noted for $(CeO_2/YBCO)_N$ multilayer films, and particularly for H < 1 T [13].

A different way of plotting $J_c(H, T)$ is shown in Fig. 3, which plots J_c as a function of temperature for $H = 3$ T and 7 T fields. The relative performance of the YBCO+nanoadditions films to YBCO can be compared there. It can be seen that it is possible to achieve $J_c = 2–3\ MA/cm^2$ (equivalent to $J_e = 20–30\ KA/cm^2$ for 100 micron thick engineered wires) with addition of optimized nanoadditions, and the operation temperature can be increased by almost 20 K on average, for both H = 3 T and H = 7 T.

Finally, the increase of J_e that could be realized with optimized nanoadditions introduced into commercial YBCO wires is plotted in Fig. 4, assuming the films can be reproduced with commercial scale, and using a standard architecture and 1 micron YBCO+nanoaddition film thickness. The improvement of J_e that could be realized is remarkably high, and increases the relative operation temperature by ~30 K to ~60 K for motor and generator applications. Also importantly, it raises the absolute value of J_e at lower temperatures which is critical for specialized applications requiring high fields H > 8 T, that might not be realized by changing the architecture.

The flux pinning properties are correlated to the film and nanoparticle microstructures, which are shown in Fig. 5. For $(M/YBCO)_N$ multilayer films, the $M = Y_2O_3$ and Y211 nanoparticles are mostly randomly oriented with size ~7–8 nm, whereas the M = BZO nanoparticles are highly oriented in layers with size ~5 nm. For $(YBCO)_{96Vol\%}(BZO)_{4Vol\%}$ films, the BZO additions organized into self-assembled nanorods with size ~8 nm. The pinning defect size is an important parameter affecting flux pinning, and it is suggested that to

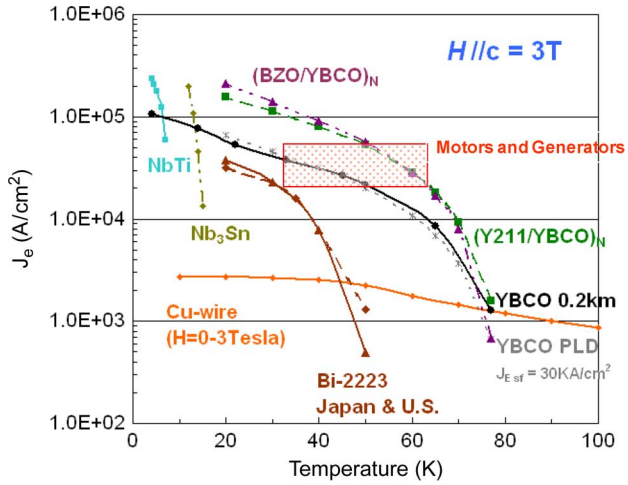

Fig. 4. Engineering current density as a function of temperature for km length commercially produced HTS and LTS wires, except YBCO which is shown for 0.2 km length [4]–[9]. The improvement of J_e possible with nanoadditions is shown for $(\mathrm{Y211/YBCO})_\mathrm{N}$ and $(\mathrm{BZO/YBCO})_\mathrm{N}$ multilayer films, assuming 1 micron film thickness, J_cs from Fig. 2 which conservatively assumes $J_{ct}(77\ \mathrm{K, self-field}) = J_{cm}(77\ \mathrm{K, self-field})$, and total film architecture 0.01 cm thickness and 0.4 cm width.

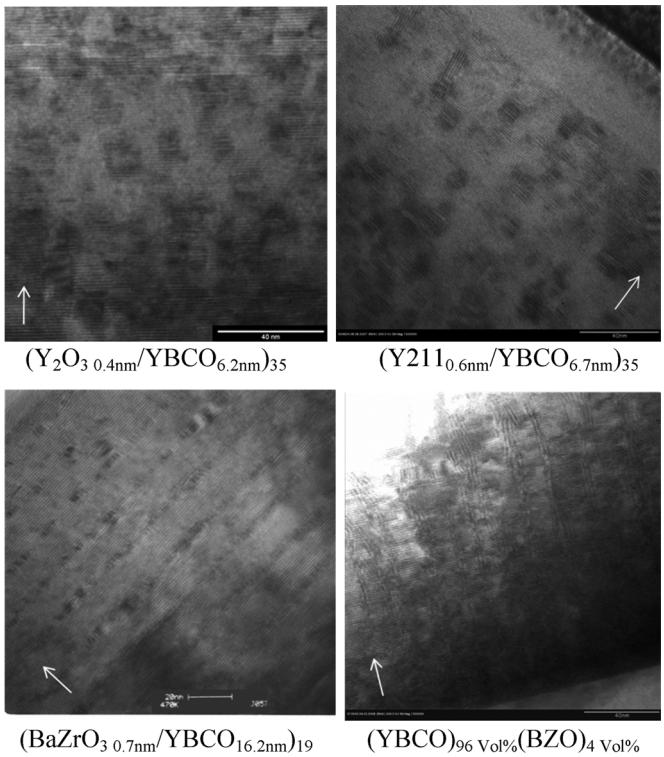

Fig. 5. Transmission electron micrographs of YBCO+nanoparticle films. Arrows indicate c-axis direction.

optimize flux pinning the defect size should be ∼2 times the coherence length, or ∼8 nm at 77 K [2], [3]. This suggests that for $(\mathrm{BZO/YBCO})_\mathrm{N}$ multilayer films, the BZO nanoparticle size might be too small to pin effectively at 77 K, but would be large enough at lower temperatures when the coherence length reduces gradually; e.g. down to ∼2 nm at 5 K.

IV. Conclusion

In summary, the $J_c(H, \mathrm{T})$ properties of varying YBCO+nanoddition films were remarkably different when compared at different magnetic fields $H = 0$–9 T and temperatures 20–77 K. One exception however was for $(\mathrm{Y211/YBCO})_\mathrm{N}$ and $(\mathrm{Y_2O_3/YBCO})_\mathrm{N}$ multilayer films which had very similar $J_c(H, \mathrm{T})$. We argue the $J_c(H, \mathrm{T})$ properties are not intuitively obvious when considered beforehand, so the question is posed about what causes these differences in $J_c(H, \mathrm{T})$. However since modeling of $J_c(H)$ at any temperature is not clearly understood yet in terms of flux pinning models, the answer is still unknown. One simple explanation for the unusual temperature dependence of $(\mathrm{BZO/YBCO})_\mathrm{N}$ films especially at 77 K is that the BZO nanoparticle size ∼5 nm is too small to pin the vortex structure with size ∼4 nm at 77 K [3]. Further experiments are needed to study and understand the $J_c(H, \mathrm{T})$ properties for the varying YBCO+nanoaddition films.

The results here indicate that by introducing optimized flux pinning additions into commercial wire manufacturing processes, significant increases of $J_e(H, \mathrm{T})$ are expected, which would increase the operation temperature of motors and generators from ∼30 K to ∼60 K. Also importantly, addition of optimized nanoparticles would increase the absolute value of J_e that can be achieved for YBCO commercial wires without changing the wire architecture, to make YBCO increasingly competitive compared to NbTi and $\mathrm{Nb_3Sn}$ for any application where absolute high values of J_e would be desired regardless of operation temperature.

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